Autothermal Cyclic Reforming Based Hydrogen Generating and Dispensing System

Ravi Kumar (Primary Contact), Shawn Barge, Parag Kulkarni, Court Moorefield, and Vladimir Zamansky
GE Global Research
General Electric Company
18 Mason
Irvine, CA 92618
Phone: (949) 859-8851 (ext. 159); Fax: (949) 859-3194; E-mail: kumarr@research.ge.com

James Smolarek, Michael S. Manning, Safdar A. Baksh, and Joseph M. Schwartz
Praxair Inc.
175 East Park Drive
Tonawanda, NY 14151
Phone: (716) 879-2761; Fax: (716) 879-7218

Michael Jones
BP
Chertsey Road, Sunbury on Thames
Middlesex, TW16 7LN
United Kingdom
Phone: (011) 44 -1932 768265; E-mail: JonesMID@bp.com

DOE Technology Development Manager: Mark Paster
Phone: (202) 586-2821; Fax: (202) 586-9811, E-mail: Mark.Paster@ee.doe.gov
ANL Technical Advisor: Thomas Benjamin
Phone: (630) 252-1632; Fax: (630) 252-4176; E-mail: Benjamin@cmt.anl.gov

Objectives

The overall objective of the project is to develop a reformer-based hydrogen refueling station capable of delivering at least 40 kg/day of hydrogen. The specific performance objectives of the refueling system are as follows:

- Produce proton exchange membrane (PEM) fuel cell grade hydrogen (99.99+% purity);
- Achieve fully automated operation during normal, start-up, shut-down and stand-by modes; and
- Achieve 75% hydrogen generator efficiency (HHV - higher heating value basis).

Technical Barriers

This project addresses the following technical barriers from the Hydrogen Production section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year R,D&D Plan:

- A. Fuel Processor Capital Costs
- B. Operation and Maintenance (O&M)
- C. Feedstock and Water Issues
- E. Control and Safety
- Z. Catalysts
- AB.Hydrogen Separation and Purification

Approach

- Design a pre-commercial 40 kg/day hydrogen generating and refueling system to produce fuel cell grade hydrogen from natural gas (NG) based on GE's Autothermal Cyclic Reforming (ACR) process;
- Analyze several process configurations that include ACR reactor, shift reactor, pressure swing adsorber (PSA), and heat exchangers and select the best configuration that has high efficiency, high reliability and lower capital cost;
- Fabricate and operate the ACR-based hydrogen generator;
- Develop a control system for safe operation of the hydrogen generator with low operation and maintenance (O&M) cost; and
- Develop tools to quantify the efficiency, cost and reliability of the system.

Accomplishments

- Assessed the technical feasibility of the design;
- Defined system layout;
- Designed ACR reactors, shift reactors and PSA;
- Operated low-pressure ACR and shift reactors for extended periods of time;
- Operated PSA on simulated reformate;
- Determined the delivered cost of hydrogen based on ACR technology; and
- Determined the competitiveness of the design relative to alternative concepts.

Future Directions

- Modify low-pressure reformer design to operate at high pressure;
- Integrate high-pressure ACR system with Praxair's PSA, hydrogen compressor, and storage system; and
- Safely install and operate the refueling system at a demonstration site.

Introduction

GE is developing a hydrogen generation system designed for vehicle refueling. The hydrogen generation system uses a proprietary reformer to convert hydrocarbon fuels to a hydrogen-rich gas that is purified downstream. The ACR process is a unique technology that can be applied for the production of hydrogen or syngas from different fuels, including natural gas, diesel fuel, and renewable feed-stocks, such as bio-derived fuels. The refueling system also includes a PSA unit to purify the hydrogen, a hydrogen compressor, high-pressure storage tanks, and a dispensing unit to safely deliver the hydrogen from the storage tanks to the vehicle. Praxair will develop the PSA unit. They will also procure the hydrogen compressor, hydrogen storage tanks, and hydrogen dispenser. BP will analyze the refueling station logistics and safety.

ACR is an autothermal cyclic catalytic steam reforming technology for converting hydrocarbons to a hydrogen-rich stream. The ACR process operates in a three-step cycle that involves steam reforming of the fuel in a Ni catalyst bed (Step 1 - Reforming), heating the catalyst bed through oxidation of the Ni catalyst (Step 2 - Air Regeneration), and finally reducing the catalyst to the metallic state (Step 3 - Fuel Regeneration). The heat required for the endothermic reforming step is provided during the exothermic air regeneration step. The ACR process consists of two reactors cycling between the reforming and regeneration (air and fuel) steps to produce a continuous stream of hydrogen. The reformer produces a 70% hydrogen stream that is purified downstream to achieve PEM fuel cell quality. The ACR process represents a significant technological advancement in comparison with autothermal reforming (ATR) and partial oxidation (POX), as the ACR-produced syngas is not diluted with nitrogen and the overall efficiency of the ACR process is higher than that of ATR and POX. When compared to conventional steam methane reforming (SMR), the ACR process has significantly lower capital costs and lower emissions. In addition, the ACR process is fuel flexible and has been successfully demonstrated using high-sulfur fuels.

Approach

The major goal of the ACR-based hydrogen generation and dispensing system project is to deliver PEM fuel cell grade hydrogen for vehicle refueling at the hydrogen cost target of $2.50/kg. The ACR technology promises to reduce the capital cost and improve the efficiency and reliability of the reformer when compared to other reforming technologies (SMR, ATR and POX). The project is broken down into three phases: Phase I - Conceptual Design and Analysis, Phase II - Sub-System Development, and Phase III - Prototype Design, Fabrication, and Operation.

In Phase I, a conceptual design of the entire ACR-based refueling system was developed. The mass and energy balances, process flow diagrams and systems design for the ACR reactors and other components were completed. A market analysis was performed to determine the competitiveness of the design relative to alternative concepts. Finally, an economic analysis of the system was performed to determine if the DOE targets are attainable using ACR technology.

Phase II is sub-system development. The major task in this phase is ACR reactor and catalyst development. The ACR catalyst will be subject to detailed evaluation for fuel conversion efficiency and reliability under different operating conditions. Phase III is prototype design, fabrication and system operation. In this phase, the entire system, including the reformer, PSA, hydrogen compressor, and storage tanks, will be integrated, installed and operated at a demonstration site.

Results

The conceptual design of the ACR system has been finalized. Multiple configurations were evaluated from a standpoint of efficiency, reliability, and capital cost. One major design issue was whether to use high-pressure reforming (150 psig) or low-pressure reforming (5 psig) with a syngas compressor. It was determined that high-pressure reforming was both more efficient and cost effective than low-pressure reforming, as shown in Table 1.

Table 1. Comparison of High-Pressure Reforming and Low-Pressure Reforming

	High-Pressure Reforming (150 psig)	Low-Pressure Reforming (5 psig)
Efficiency (LHV)	75%	71%
Capital Cost	Lower	Higher
Reliability	Higher	Lower, due to syngas compressor

The layout of the selected system is shown in Figure 1. The major subsystems of the refueling system are reformer, shift reactor, PSA, hydrogen compressor, storage tanks, and dispenser. The station will be capable of refueling three vehicles consecutively. The footprint of the refueling system was evaluated. The largest components in terms of size are the storage tanks.

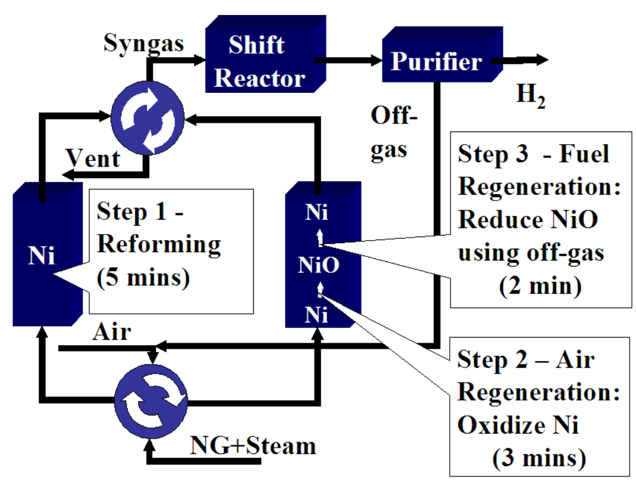

Figure 1. Refueling System Layout

A low-pressure pilot-scale reformer was tested for extended periods of time. The reformer was subjected to several start-stop cycles and operated using automated controls. Figure 2 shows that the reformer operates in a stable manner, and the reformate has about 70% hydrogen.

Praxair has determined the optimal configuration of the PSA for the ACR cyclic process. The PSA will be a 3-bed system designed for low capital cost as well as easy valve maintenance. Praxair performed an extensive evaluation of hydrogen compressors based on capital cost, reliability, and the ability to meet the performance targets. They chose a hydraulically driven compressor. One of the main factors for this choice was its oil-free design that will help prevent contamination of the hydrogen. The compressor has a long, slow stroke that will result in higher reliability. The design allows for quick maintenance that will decrease O&M cost for the system. Praxair is considering both steel and composite tanks for the hydrogen storage system. The major factors in this selection are safety and codes. Multiple methods to fill the vehicle were evaluated, and it was concluded that a cascade system was optimal.

A detailed cost analysis of the entire refueling station was performed to estimate the cost of delivered hydrogen. The model takes into account the system capital cost, O&M cost, and fuel and consumables costs. Figure 3 shows the cost of delivered hydrogen at different system capacities and mass production rates. The plot shows that the refueling system can meet the DOE target at higher capacities (>20,000 scfh) and when mass-produced (>500 units/year).

A sensitivity analysis was performed on the factors that affect the cost of delivered hydrogen (see Figure 4). The factors that were varied were: NG cost, O&M cost, capital cost, efficiency, and availability. Each of these factors were varied one at a time. Figure 4 shows the cost of delivered H2 at the mean value for each of the factors (center points), as well as the variability in the cost of H2 due to the variability in each of the factors. For example, as shown in Figure 4, the cost of NG has a mean of $4/

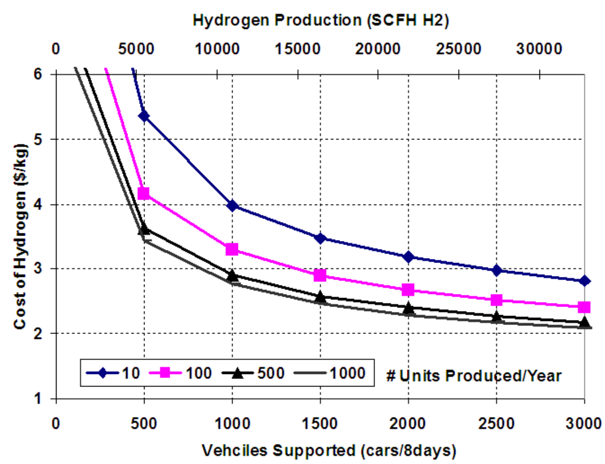

Figure 3. Cost of Hydrogen Estimated for Various System Sizes and Mass Production Rates

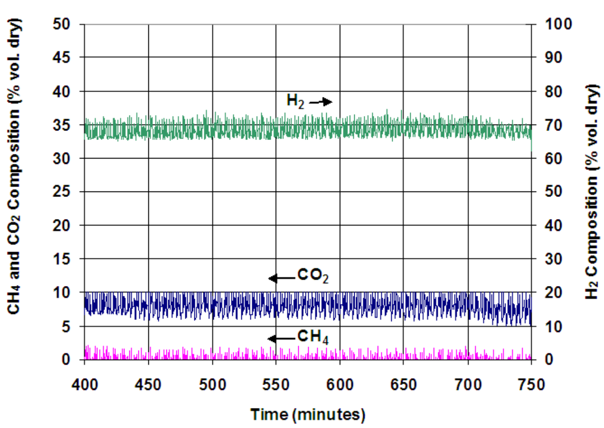

Figure 2. Performance of ACR

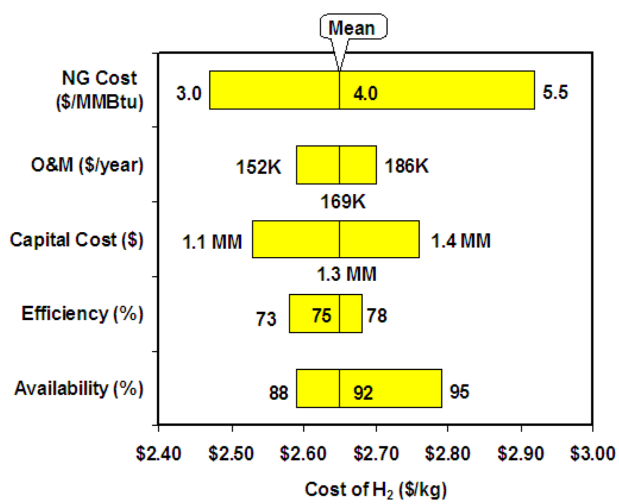

Figure 4. Sensitivity Analysis of Cost of Hydrogen

MMBtu and a variability of \$3.50-\$5.00/MMBtu. This results in a delivered cost of H2 with a mean of \$2.65/kg and a variability of \$2.47-\$2.92/kg. The analysis determined that the variability in cost of hydrogen will be primarily due to variability in NG cost, capital cost, and availability.

Conclusions

The process design of the reformer, shift reactor, and PSA has been completed. The design was optimized to increase efficiency, decrease capital costs, and improve reliability.

The process and economic analysis determined that high-pressure reforming is better than low-pressure reforming for the ACR-based hydrogen generator. A low-pressure reformer was operated for extended periods using automated controls. The reformer operated in a stable manner and delivered a 70% hydrogen stream to the shift reactor. The design of the low-pressure reformer is being modified to allow the reformer to operate at high pressure. The design of the shift reactor has been completed.

After extensive analysis, a three-bed PSA design was chosen over other bed designs in order to reduce capital costs as well as to reduce operation and maintenance costs. Vendors for hydrogen compressor, storage tanks, and dispenser have been identified. Hydro-Pac was chosen for the hydrogen compressor because of their unique slower speed hydraulic drive unit and successful prior experience with Praxair in high-pressure gaseous nitrogen and argon compression. Praxair is considering both steel and composite tanks for the hydrogen storage system. Fueling Technologies Incorporated has been chosen as the vendor for the dispenser. Multiple methods to fill the vehicle were evaluated, and it was concluded that a cascade system is optimal.

The economic analysis indicated that the refueling system will meet the hydrogen cost target of \$2.50/kg at capacities greater than 20,000 scfh and when mass-produced in greater than 500 units/year.

FY 2003 Publications/Presentations

1. Hydrogen Refueling System Based on Autothermal Cyclic Reforming, Kumar, R., International Energy Agency Annexe XV Meeting, Palm Springs, CA, November 17-18, 2002.

2. Hydrogen Refueling Station Based on Autothermal Cyclic Reforming, Kumar, R., Barge, S., Kulkarni, P, Moorefield, C, Zamansky, V., Fuel Cell Seminar, Palm Springs, CA, November 18-22, 2002.